

Olson and Lynch Reply: The accompanying Comment [1] reports new measurements on Eu films on Ta(110) in which an apparently insulating phase we reported earlier [2] was not observed. It reports new calculations showing that fcc and hcp Eu have no band gap. It suggests the gap may have been due to the presence of hydrogen, i.e., the films may have been EuH_2 , an insulator.

We cannot criticize these new results; Wahl *et al.* obtained spectra that are reasonable, and their calculation shows what may be expected for a divalent metal, a low density of states at the Fermi level, or a small gap. Our previous results were reproduced many times in three different UHV chambers, with several samples of source Eu. The predominant residual gas in UHV systems is hydrogen, undetected by Auger or x-ray photoemission spectroscopy spectroscopies (which ruled out an oxide). The pellets of Eu were outgassed extensively, but we always found one of two photoemission spectroscopy (PES) spectra, depending on substrate, the one reported and one for bcc Eu. We also made Eu films with the chamber partially backfilled with H_2 but saw no difference from films prepared at typically 3×10^{-10} Torr [2]. Unfortunately, the absence of H in or on the films is not proven. The plasmon energy was found in both phases to be near that of bcc Eu, significantly different from that estimated for EuH_2 . (We later found electron energy loss (EEL) data on several rare-earth dihydrides (not EuH_2) that place the hydride plasmon about 2 eV higher than that of the rare earth [3].) Our EEL measurements used 800 eV electrons, which sample a greater depth than sampled in valence-band photoemission. This suggests that any H present could be on the surface of the new phase, but not in the bulk. This is difficult to demonstrate with the probes used. However, when the close-packed films reverted to the bcc phase, there was no spectral contribution in the 4–5 eV region. Those “reverted” films had the same PES spectra as films prepared directly in the fcc phase. If H were present in the PES sampling depth of several monolayers in the amount required if the spectrum of the close-packed phase were that of a dihydride, it quickly either desorbed or dissolved in the bcc phase when the phase changes. Hydrogen is not very soluble in bcc Eu, but its diffusion rate is high [4]. If all of the H in, e.g., three monolayers (ML) of surface EuH_2 were to dissolve into the 10–20 monolayers of bcc Eu that appeared when the film thickness became too great, it would exceed the bulk solubility limit. However, the epitaxial bcc films have domains, and H could migrate to the domain boundaries. Thus we cannot rule out a surface hydride as the insulating phase followed by H at domain boundaries in the bcc phase.

We were concerned about the spectrum of the insulating phase in the 4–5 eV region because no calculation showed Eu 5d states at such large binding energies. Photoelectron spectra were taken with a wide range of photon energies. The 4–5 eV region had increased strength at the low

photon-energy side of the resonance peak, suggestive of 5d character. The spectra were not conclusive because the 5d cross section is much less than that of the 4f at these photon energies [5], and the structure in question is in the tail of the resonant 4f emission and the associated secondaries.

The lattice parameters of the films were quite reproducible, allowing for errors in locating spots on the printed low energy electron diffraction (LEED) patterns and in thicknesses of different films. From LEED patterns the minimum interatomic spacing was 4.06 Å at about 1 ML of Eu. The maximum spacing was 4.5–4.6 Å at the initial appearance of the hexagonal LEED pattern, and again at coverages above 3 ML. Figure 3 in Ref. [2] is labeled “10 Å,” from a quartz thickness monitor. (The Eu film condensed on the monitor is probably bcc.) The thickness of the corresponding epitaxial layer is not known. The nearest-neighbor spacing of 4.06 Å results from the figure. A still thicker film gave a smaller hexagonal LEED pattern, rotated a few degrees with respect to that of Fig. 3. We agree that 4.5 Å is a surprisingly large nearest-neighbor spacing. EuH_2 is normally orthorhombic [6]. GdH_2 has the CaF_2 structure, with Gd atoms in a close-packed array on 111 planes. Their nearest-neighbor spacing is 3.56 Å [6]. The shortest Gd-H distance is 2.18 Å, not in this plane. Both distances are significantly smaller than those we observed.

We cannot dispute the results of Wahl *et al.* [1]. Their suggestion that the insulating phase we reported in [2] is due to H cannot be proven or disproven with the evidence at hand. But a surface layer of a hydride as thick as the PES sampling depth cannot be ruled out. Where the H goes when the close-packed phase reverts to bcc is not known. A more convincing case for the hydride could be made if another group could reproduce the films we obtained by incorporation of H.

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